

# Final Report

Composite-Based High Performance Electroactive Polymers  
For Remotely Controlled Mechanical Manipulations in NASA Applications

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## **Abstract**

This program supported investigation of an all-polymer percolative composite which exhibits very high dielectric constant ( $>7,000$ ). The experimental results show that the dielectric behavior of this new class of percolative composites follows the prediction of the percolation theory and the analysis of the conductive percolation phenomena. The very high dielectric constant of the all-polymer composites which are also very flexible and possess elastic modulus not very much different from that of the insulation polymer matrix makes it possible to induce a high electromechanical response under a much reduced electric field (a strain of 2.65% with an elastic energy density of  $0.18 \text{ J/cm}^3$  can be achieved under a field of 16 MV/m). Data analysis also suggests that in these composites, the non-uniform local field distribution as well as interface effects can significantly enhance the strain responses. Furthermore, the experimental data as well as the data analysis indicate that the conduction loss in these composites will not affect the strain hysteresis.

## 1. Publications and presentations

- Cheng Huang and Qiming Zhang. Enhanced Dielectric and Electromechanical Responses in High-dielectric-constant All-Polymer Percolative Composites. To be published in *Advanced Functional Materials* (2004).
- Cheng Huang, Ji. Su, and Q. M. Zhang. High dielectric constant all-organic/polymer composite actuator materials. *Proc. Symp. Mater and Devices for Smart Systems of MRS 2003 Fall meeting*, MRS Vol. 785 (Boston, 2003)
- Q. M. Zhang, Cheng Huang, Feng Xia, and Ji Su. Electric EAP, Chapter 4 in *Electroactive Polymer (EAP) Actuators as Artificial Muscles*, Y. Bar-Cohen, ed. (SPIE, WA, to be published in 2004).
- Feng Xia, Hengfeng Li, Cheng Huang, M. Y. M. Huang, H. Xu, Z.-Y. Cheng, and Q. M. Zhang. P(VDF-TrFE) Based High Performance Electroactive Polymers. *Proc. of SPIE Symposium on Electroactive Polymer Actuators and Devices*, Vol. 5051 (SanDiego, CA, 2003).
- Cheng Huang, Hengfeng Li, Feng Xia, Z. Y. Cheng, and Q. M. Zhang. Development of High Electromechanical Response Polymers. First World Congress on Biomimetics and Artificial Muscles, Albuquerque, Dec. 9-11, 2002.
- Q. M. Zhang. Advanced Ferroelectric Polymers for Artificial Muscles. First World Congress on Biomimetics and Artificial Muscles, Albuquerque, Dec. 9-11, 2002 (Invited)
- Cheng Huang, Ji. Su, and Q. M. Zhang. High dielectric constant all-organic/polymer composite actuator materials. MRS 2003 Fall meeting, Boston Dec. 1-Dec. 4, 2003.

## 2. Introduction

In this program, we investigated an all-polymer percolative composite with the aim of achieving high dielectric constant. As has been shown in several recent publications,<sup>[1]</sup> raising the dielectric constant of a polymeric material is one of the key factors in the electroactive polymers to realizing much reduced operation field in generating high field-induced strain while maintaining high elastic energy density. In addition, a polymeric material with high dielectric constant is also attractive for charge storage and other capacitive applications.

In recent years, several electroactive polymers, which responses are controlled by external electric fields, hereafter referred to as the field effect EAPs, have been developed or discovered to exhibit high field-induced strain (>5% strain) with high elastic energy density ( $\sim 1 \text{ J/cm}^3$ ).<sup>[2-4]</sup> However, one severe drawback of these EAPs is the very high operation fields (>100 MV/m) required to generate such a high strain with high elastic energy density,

which limits these EAP applications, especially in biomedical fields. As has been pointed out in an earlier publication,<sup>[1]</sup> one fundamental cause for such high fields is the energy conservation, i.e., the elastic energy density generated in an electroactive polymer can't exceed the input electric energy density, which is  $K\epsilon_0 E^2/2$  for a linear dielectric where  $K$  is the dielectric constant,  $\epsilon_0=8.85 \times 10^{-12}$  F/m is the vacuum permittivity, and  $E$  is the applied field, if there is no other energy source available in the material. For example, even to generate an elastic energy density of  $0.1 \text{ J/cm}^3$ , which is the value for the best performed piezoelectric ceramics,<sup>[2]</sup> and assuming 50% energy conversion efficiency, the field required is  $E=95 \text{ MV/m}$  for an electroactive polymer with a dielectric constant of 5, which is a typical value for most electroactive polymers currently developed. This simple analysis points out the importance of raising the dielectric constant of the electroactive polymers to achieve high electromechanical responses.

For traditional polymeric materials, it is well known that the dielectric constant is relatively low ( $<10$ ) in comparison with those in the inorganic materials many of which can exhibit dielectric constant higher than 5,000. It is noted that conductive percolation phenomenon in composites has been studied for many decades.<sup>[5,6]</sup> In most of the early studies, it is the conductive properties of the composites that are investigated. Several recent experiments have demonstrated that the percolative phenomenon can also be utilized to enhance the dielectric response of the composites.<sup>[7,8]</sup> In composites of conductive fillers embedded in an insulation matrix, the variation of the dielectric constant (both the real and imaginary parts) of the composite with the concentration  $f$  of the conductive filler has been predicted to follow a critical behavior,<sup>[6,9]</sup>

$$K = K_m \left\{ \frac{f_c - f}{f_c} \right\}^{-q} \quad (1)$$

where  $K_m$  is the dielectric constant of the insulation matrix,  $q$  is a critical exponent ( $\sim 1$  for a three dimensional composite), and  $f_c$  is the percolation threshold for the conduction. Equation (1) indicates that as  $f$  approaches  $f_c$ , a large dielectric enhancement can be obtained. Further more, to achieve a high dielectric constant, a matrix with a relatively high dielectric constant is also highly desirable.

The main objective of this work is to raise the dielectric constant of a polymeric material substantially above the current level so that it can generate high electromechanical

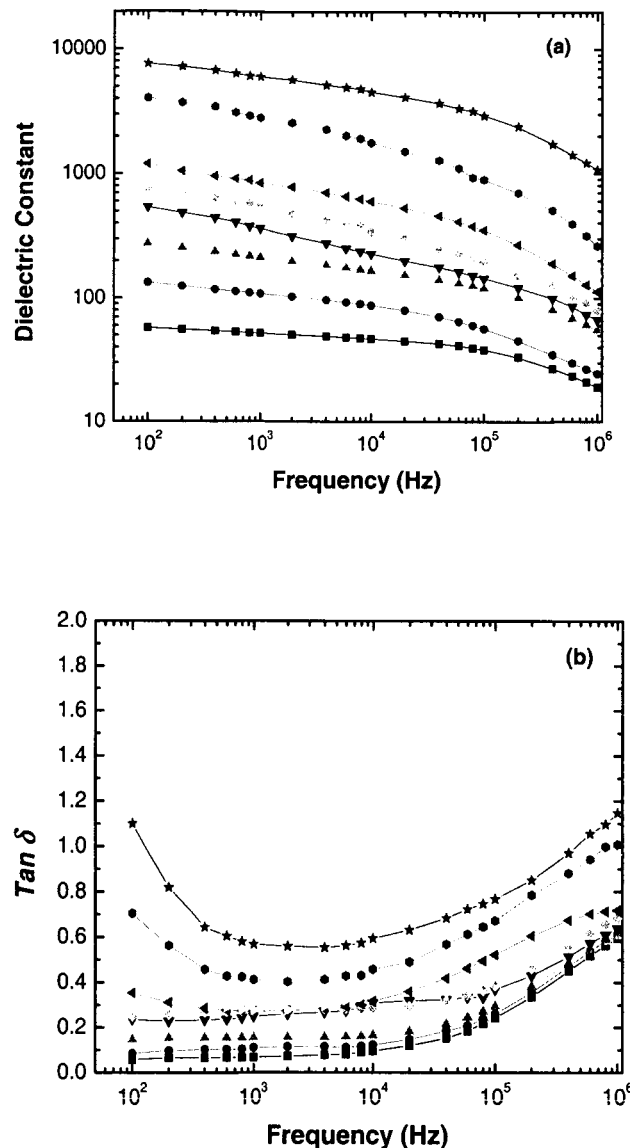
responses with a much reduced applied field level. It is obvious that in addition to electromechanical applications, such a polymeric material is also attractive for all organic electronics.

### 3. Results and Discussion

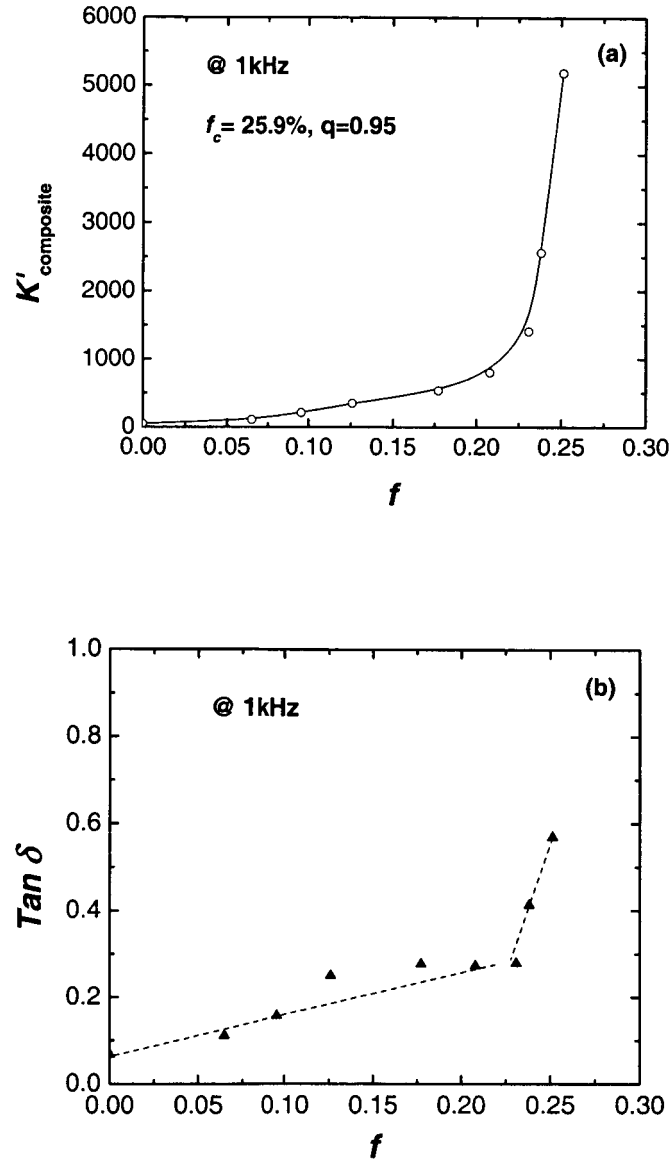
For the percolative composites investigated here, the poly(vinylidene fluoride-trifluoroethylene-chlorotrifluoroethylene) (P(VDF-TrFE-CTFE)) 62/26/12 mol% which is a relaxor ferroelectric polymer, possessing a relatively high room-temperature dielectric constant ( $\sim 50$ ) and a relatively high electrostrictive strain response.<sup>[10]</sup> As shown in equation (1), a high matrix dielectric constant is a necessary condition to achieve very high dielectric constant for the percolative composites even when the conductive filler concentration  $f$  is not very close to  $f_c$ . In addition, a high electromechanical response in the insulation matrix is also highly desirable for the composites to realize high electromechanical responses under a much reduced electric field. The conductive polymer PANI as the conductive filler, which has a relatively low elastic modulus ( $\sim 2.3$  GPa) in comparison to metal particles, will not change the modulus of the composite very much from that of the polymer matrix.

Presented in Figure 1 is the dielectric constant and loss for the composites with different volume concentration of the PANI as a function of frequency measured at room temperature. For the composite with  $f=0.251$ , the dielectric constant reaches more than 7,000 and the loss tangent is at 0.6 when measured at 1 kHz. Summarizing the data in Figure 1 (real part of the dielectric constant) at 1 kHz as a function of  $f$ , the conductive phase volume concentration in the composites, yields the data in Figure 2. Fitting the data in Figure 2 to equation (1) yields an  $f_c=0.259$  and  $q=0.95$ . The critical exponent  $q$  is close to that predicted for a 3-dimensional percolative system with spherical shape conductive particulates. The loss tangent shows increases as  $f_c$  is approached which could be caused by the inhomogeneity in the composites which raises the conduction in the composites above the level predicted by the percolation theory. In fact, the analysis from the percolation theory indicates that the loss tangent should not change with  $f$  at  $f < f_c$ . On the other hand, in the real composites, there exists small composition variation and these sub-phases can form parallel path which behavior can affect the imaginary part of the dielectric constant quite significantly as the composition approach  $f_c$ , which we believe is the reason that causes the large increase in the

dielectric loss at near  $f_c$ . Similar phenomenon has also been observed in the inorganic percolative composites.<sup>[7,8]</sup> We will come back to this point when we discuss the data of the field induced strain.



**Fig. 1.** The weak-field (150V/cm) dielectric constant and dielectric loss as a function of frequency measured at room temperature for the composites with different volume concentration  $f$  of the PANI, compared with the P(VDF-TrFE-CTFE) matrix. (volume concentrations  $f = 0, 0.065, 0.096, 0.127, 0.177, 0.208, 0.230, 0.238$ , and  $0.251$  from the bottom to top for the dielectric constant (a) and the loss (b)). The data points are shown and solid curves are drawn to guide eyes.



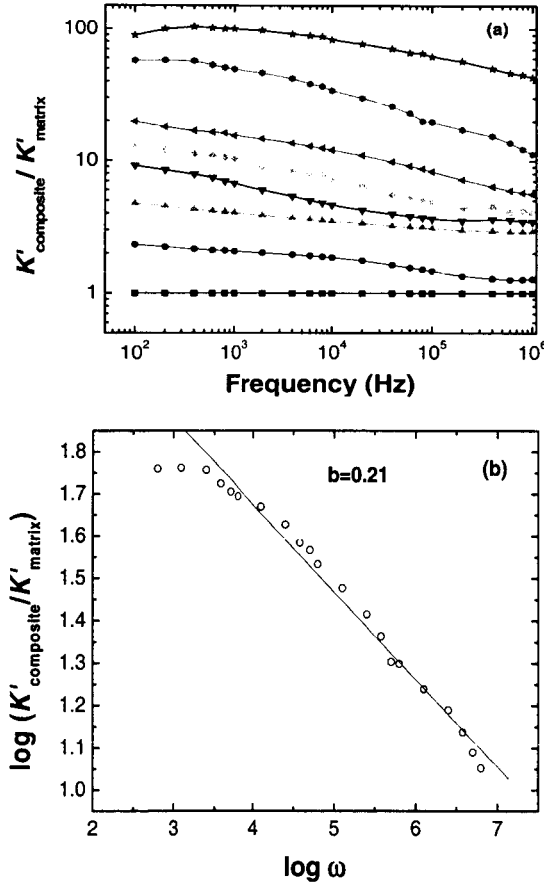
**Fig. 2.** a) Evolution of the real part of the dielectric constant of the composites versus the PANI volume concentration,  $f$ , at the room temperature measured at a frequency of 1 kHz. The continuous line represents the fit of the experimental points to Equation (1). b) Dependence of the dielectric loss versus  $f$  measured at a frequency of 1 kHz. The dashed line is a guide for the eyes.

The data in Figure 1 also reveal that the dielectric properties of the composites change with frequency. This frequency dispersion is caused by two main factors. One is from the frequency dispersion of the dielectric matrix (the data in Figure 1 for  $f=0$ ) and the other is due to the composite nature of the material. Several analyses indicate the power law

frequency dispersion for both the real ( $K'$ ) and imaginary ( $K''$ ) part of the dielectric constant of the composites:<sup>[6,11,12]</sup>

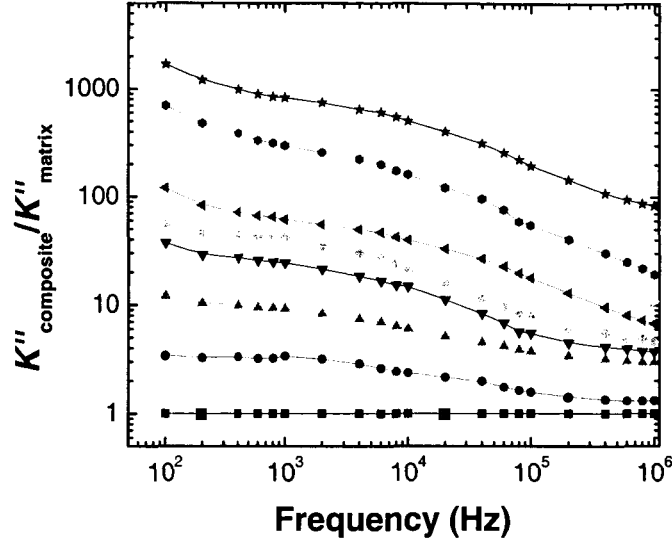
$$K' \sim \omega^{-b} \text{ and } K'' \sim \omega^{-b} \quad (2)$$

where  $\omega$  is the angular frequency and  $0 < b < 1$ . To remove the dispersion from the polymer matrix, the ratio of the composite dielectric constant to the matrix dielectric constant is used in the data analysis (Figure 3 (a) and Figure 4). The fitting of  $K'$  yields an exponent “ $b$ ” ranging from 0.14 to 0.21 (Figure 3(b) for the composite with  $f=0.238$ ). Due to the conduction in the composite originating from the inhomogeneity in the composites which can affect markedly the imaginary part of the dielectric constant, no data analysis was performed on  $K''$  (imaginary part of the dielectric constant).



**Fig. 3.** a) Frequency dependence of the real part ratio of the composite dielectric constant to the matrix dielectric constant for the composites with different volume concentration of the PANI,  $f = 0, 0.065, 0.096, 0.127, 0.177, 0.208, 0.230, 0.238$ , and  $0.251$  from the bottom to top. b) Variation of  $\log K'$  vs.  $\log \omega$  for the composite with  $f=0.238$ . The straight line has a slope of 0.21.





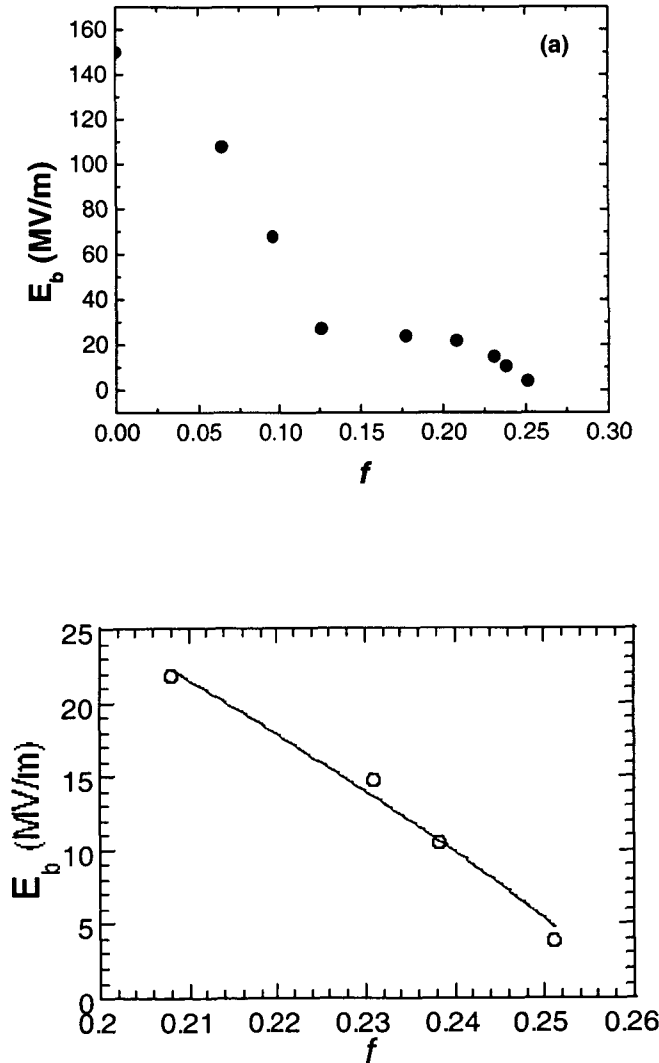
**Fig. 4.** a) The ratio of the imaginary part of the composite dielectric constant to the matrix dielectric constant as a function of frequency for the composites with different volume concentration of the PANI,  $f = 0, 0.065, 0.096, 0.127, 0.177, 0.208, 0.230, 0.238,$  and  $0.251$  from the bottom to top.

The dielectric strength (breakdown field) was measured for these composites and the data are presented in Figure 5. The breakdown field decreases continuously from the pure P(VDF-TrFE-CTFE) matrix ( $\sim 160$  MV/m) to  $3.85$  MV/m when  $f=0.251$ . For percolative composites, as  $f$  increases, it is expected that there are regions in the composites at which the insulation layer thickness  $d$  between the conductive particles decreases which causes the increase of the effective dielectric constant of the whole composites. In these regions, the local fields will increase, implying that the breakdown field will decrease with  $f$ . Indeed, a model analysis has shown that for a 3-dimensional percolative composite, the breakdown field follows

$$E_b \sim (f_c - f)^s \quad (3)$$

where  $s=1$ .<sup>[13]</sup> On the other hand, for pure dielectric films, many experimental evidence as well as theoretical models show that the breakdown field  $E_b$  will increase as the film thickness  $d$  is reduced although different models predict different thickness dependence behavior.<sup>[14,15]</sup> In some polymer films,  $E_b$  is approximately proportional to  $(a + d^{-\gamma})$ , where  $\gamma$  is less than 1 and  $a$  is a constant.<sup>[16]</sup> Combining the two effects yields that the breakdown field in the percolative composite films may vary as  $(f_c - f)^p$  where the exponent  $p$  is less than  $s$ . It should also be noted that for the composites studied here, the presence of defects and

impurities (for example, residual solvent and trapped air bubbled air) is also an important cause of electric breakdown. In addition, the possible aggregation of the PANI in the composites will affect the dependence of the breakdown field on  $f$ , the volume concentration of the PANI in the composites. For the data in Figure 5(a), the last four data points which are close to the percolation threshold are used to fit equation (3) as shown in figure 5(b) where  $f_c=0.259$  is fixed. The fitting yields the exponent  $p = 0.82$ , which is smaller than  $s=1$  and seems to be consistent with the analysis presented.



**Fig. 5.** (a) Breakdown field  $E_b$  versus the PANI volume concentration,  $f$ . (b) Breakdown field  $E_b$  as a function of  $(f_c - f)$ , where  $f_c = 0.259$ . The line is a fitting to equation (3) with  $p = 0.82$ .

The field induced strain response for the composites was characterized. It was found that at each measuring field, the strain is proportional to the square of the applied field. Figure 6 presents the amplitude of the measured strain response  $S_A$  as a function of the amplitude of the applied field  $E_A$ . Under a field of 16 MV/m (or 160 kV/cm), a thickness strain of more than 2.65% can be obtained, a marked reduction in the applied field in comparison with that in the terpolymer matrix to induce the same level of strain response (7 to 8 times). The elastic energy density at this field level is 0.18 J/cm<sup>3</sup> (the Young's modulus of the composite is 0.53 GPa) which is higher than that of the best performance piezoceramics.<sup>[2]</sup>

For the terpolymer matrix (the composite with 0% PANI), the strain amplitude also approximately depends the square of the applied field amplitude. As has been shown in early studies, the strain response in PVDF based terpolymers is electrostrictive in nature.<sup>[2,17]</sup> That is, the induced strain  $S$  is proportional to the square of the polarization  $P$  induced, i.e.,  $S = QP^2$ , where  $Q$  is the charge related electrostrictive coefficient. Assuming a linear relationship between  $P$  and  $E$ , the strain  $S$  is related to the applied field  $E$  as  $S = ME^2$  where  $M$  is proportional to the square of the dielectric constant ( $M=(\epsilon-1)^2Q$ ).<sup>[17]</sup>

For the composite system investigated here, the strain response is mainly from the P(VDF-TrFE-CTFE) matrix through the electrostrictive effect and Maxwell stress effect. For a dielectric medium under external fields, the Maxwell stress, which is due to electrostatic forces, has been derived as

$$T_{ij}=E_iD_j-1/2\delta_{ij}D_kE_k \quad (4)$$

where  $\delta_{ij}=1$  when  $i=j$ ,  $D_i$  is the electric displacement component and  $E_i$  is the electric field component, respectively.<sup>[18]</sup> For the isotropic medium studied here, the Maxwell stress induced thickness strain, assuming a uniform medium, is

$$S= - 1/2 K\epsilon_0E^2(1+2\nu)/Y \quad (5)$$

where  $K$  is the relative dielectric constant,  $\epsilon_0$  is the vacuum permittivity,  $Y$  is the elastic modulus, and  $\nu$  is the Poisson's ratio. For a non-homogeneous medium, equation (4) can still be used except that now all the quantities are the local material properties.

The elastic modulus  $Y$  of the composites was measured at room temperature at the frequency of 1-2 Hz. The elastic modulus of the composite with 23 % PANI is 535 MPa. Compared with the elastic modulus of PANI fillers of 2.3GPa and the P(VDF-TrFE-CTFE)

terpolymer of 400 MPa, the filler influences the elastic properties of the composite slightly and the composites remain very flexible. For the composites with 12.7 % PANI, the modulus is about 470 MPa.

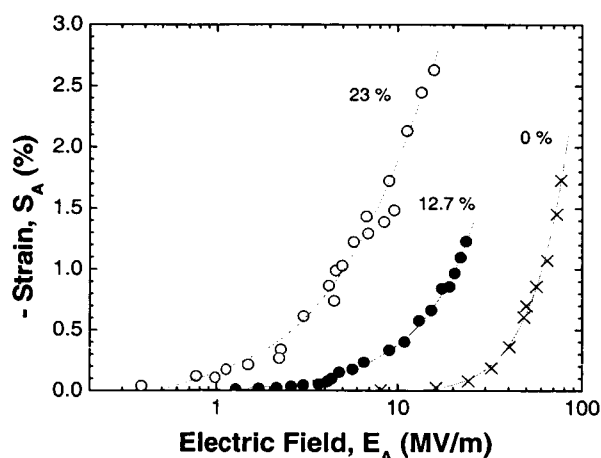
For the composites with 12.7 % of PANI, Figure 6 shows a strain of -1.25% induced under a field of 24 MV/m. With such a low PANI volume concentration, the average field in the terpolymer matrix should be nearly the same as 24 MV/m and as shown by the strain curve in Figure 6, an induced strain in the pure terpolymer, which is used as the matrix for the composites, should be -0.075% under this field. Taking the data of the dielectric constant and elastic modulus, it can be estimated from eq. (5) that a Maxwell stress induced strain accounts for -0.87% thickness strain if a Poisson's ratio of 0.4 is assumed.<sup>[19]</sup> The two strains combined accounts for more than 75% of the total observed strain response for the composite with 12.7 % of PANI. On the other hand, for the composite with 23 % of PANI, assuming a uniform field in the sample, there is very little electrostrictive strain from the terpolymer matrix under a field of 16 MV/m (see the curve in Figure 6 measured from the terpolymer matrix, where an electrostrictive strain of -0.023% is induced under 16 MV/m field), and the Maxwell stress contribution can be estimated to be about -1.33% strain ( $\nu=0.4$  is used). Therefore, measured strain response of -2.65% under 16 MV/m is approximately two times of the combined strain responses (electrostriction and Maxwell stress effect) assuming a uniform field distribution in the film. The relatively low volume concentration (23 %) of PANI in the composite makes it unlikely that strain change in PANI is responsible for the observed strain response. Combining the data from the two composites suggest that as the filler concentration increases, the strain enhancement effect (the measured strain is much larger than the strain responses estimated assuming a uniform field distribution and no-interface effect) increases. When the PANI concentration increases, it is anticipated that the field distribution in the composites will become increasingly non-uniform and interface effects from the PANI-matrix interface will also become increasingly important, which may result in an enhanced electromechanical response. Such a strain enhancement effect has also been observed in other polymer systems with heterogeneous morphologies and has been demonstrated in a recent theoretical model.<sup>[20-22]</sup>

It should be noted that for the two composites from which the strain responses were characterized as shown in Figure 6, there are large dielectric losses at 1-2 Hz frequency

range. For the composites with  $f=0.23$ , the loss tangent measured at 1 Hz at high fields is higher than 2 ( $D=\tan \delta > 2$ ). One of the questions is how this high dielectric loss affects the strain response, especially the strain hysteresis. Taking the electrostrictive strain as an example, if the complex dielectric constant  $K$  possesses a large phase angle (a high dielectric loss), the strain-field relationship will exhibit a large hysteresis due to the lagging of the strain response to the applied field (see figure 7(c)). On the other hand, if the high dielectric loss is caused by the conduction in the composites (as shown schematically in Figure 8) as likely to be the case for the composites investigated here, it will not affect the strain response and strain hysteresis. For the case shown in Figure 7, the total dielectric response of the combined material system is equal to the part  $\epsilon_1^*$  from the capacitor component, which includes various dielectric contributions, and the part  $j\sigma/\omega$  which is from the conduction through the resistor part, i.e.,

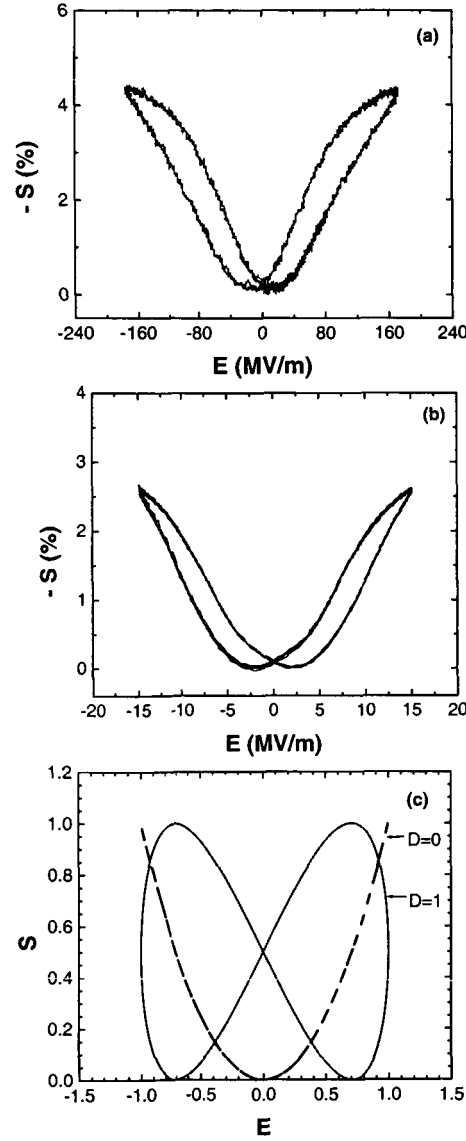
$$\epsilon^* = \epsilon_1^* - j\sigma/\omega \quad (6)$$

and the dielectric constant used in the expressions for the electrostriction and Maxwell stress induced strain should be  $\epsilon_1^*$  rather than  $\epsilon^*$  where the symbol “\*” indicates the complex dielectric constant. For the percolative composites, the loss part of  $\epsilon_1^*$  should not be very much different from that of the polymer matrix for  $f < f_c$ .

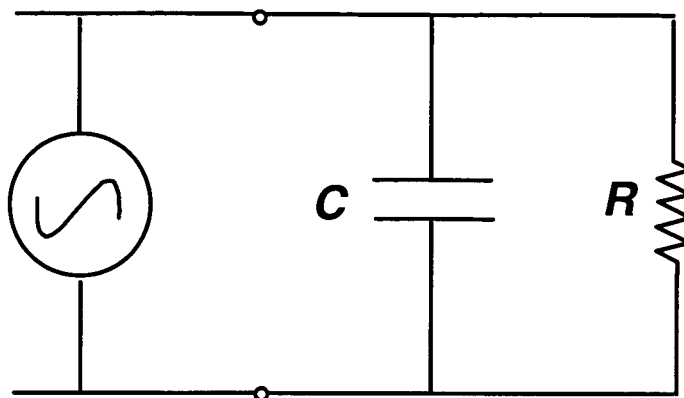


**Fig. 6.** The strain amplitude  $S_A$  as a function of the applied-field amplitude  $E_A$  measured at room temperature for the composites with  $f = 0.127$  (dot) and  $0.23$  (circle) of PANI volume concentration. For comparison, the strain from the electrostrictive P(VDF-TrFE-CTFE) terpolymer is also shown (star). The applied-field frequency is 1 Hz. Data points are shown and curves are drawn to link the data points.

Indeed, as shown in Figure 7(a) and 7(b) which compares the strain-field curve from the composite with  $f=0.23$  to that from the terpolymer matrix, apparently, the two have very similar level of hysteresis, which is relatively small (corresponds to a dielectric loss  $D = \tan \delta \sim 0.1$ ). For further comparison, we show in Figure 7(c) the strain-field curve for a hypothetical electrostrictive material with a dielectric loss  $D=1$  which is due to the dielectric relaxation. A very large strain hysteresis is observed.



**Fig. 7.** The measured strain-field curve from the composite with  $f = 0.23$  (b), compared with those from the terpolymer matrix (a), and a hypothetical material (c) with a dielectric loss  $D = \tan \delta = 0$  ( $\delta = \phi = 0^\circ$ ), and  $D = 1$  ( $\delta = \phi = 45^\circ$ ) which is from the dielectric relaxation



**Fig. 8.** An equivalent circuit for the composite near the percolation threshold where the presence of small amount of sub-phase which has compositions above the percolation forms parallel path (a resistor) with the majority phase which is still below the percolation (a capacitor), which illustrates the marked effect of the conductive minor sub-phase on the imaginary part of the dielectric constant and dielectric loss.

#### 4. Experimental

*Materials Processing:* Polyaniline was synthesized using the conventional polymerization of aniline to produce fine emeraldine hydrochloride polymer particles. <sup>[23,24]</sup> The product was extracted with ethanol to remove any unwanted impurities. The synthesized polyaniline was then coated with an insulating polymer to improve the breakdown field. Lauryl methacrylate, a long chain vinyl molecule, was chosen as the insulating monomer due to its good blend compatibility with the polymer matrix. The finely ground PANI powder was added to the degassed monomer and the reaction mixture was stirred by a magnetic stirrer and simultaneously shaken in an ultrasonic bath under a constant temperature of 60 °C. The final powder, having an average density of 1.3g/cm<sup>3</sup>, was extracted with pentanol and dried. The average particle size of coated PANI, as determined by SEM image, is below 1 μm. Fourier transform infrared (FTIR) spectra indicated that the lauryl poly(methacrylate) has bonded chemically to the PANI backbone. Composite films were prepared using the solution cast method. P(VDF-TrFE-CTFE) terpolymer powder (the average density of 1.8g/cm<sup>3</sup>) was dissolved in dimethylformamide and then a proper amount of PANI powder (determined by the volume percentage (%) of PANI in the composite) was added to the solution. The solution was ultrasonically stirred to disperse the PANI powder. After that, the solution was poured onto a glass plate and dried. The typical film thickness was about 50

μm. Gold electrodes were sputtered on the two surfaces of the films to form the electrodes for the electrical characterization.

*Dielectric and Electromechanical Measurements:* The weak-field dielectric properties of the composites were evaluated by a HP 4284 precision LCR meter in the frequency range of 100Hz to 1MHz over a broad temperature range. The high-field dielectric properties of the composites under different applied-field strength were characterized by directly measuring the magnitude and the phase of the current passing through the composite by a Lock-in amplifier under a given AC voltage. From the complex electric impedance  $Z^* = 1/(j\omega C^*)$  and  $C^* = (K' - j K'') \epsilon_0 A/t$ , the dielectric constant can be determined, where  $A$  is the area and  $t$  is the thickness of the capacitor,  $\epsilon_0$  is the vacuum permittivity ( $=8.85 \times 10^{-12}$  F/m) and  $\omega$  is the angular frequency. The elastic modulus was determined using a commercial dynamic mechanical analyzer (TA Instruments, DMA2980). The electric-field-induced strain along the applied-field direction (longitudinal strain) was measured using a cantilever-based sensor.<sup>[25]</sup> It was found that for a given field, the strain is proportional to the square of the applied field. The breakdown field was measured using a sphere to plane electrode system.<sup>[14]</sup>

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